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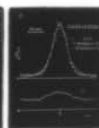
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Using the Two-Quantum Photoelectric Effect**

by

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Picosecond Time-Interval Measurement and Intensity Correlations
Using the Two-Quantum Photoelectric Effect

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We have been interested in the physical problems involved in precise time-interval measurement in the picosecond regime using mode-locked laser pulses. Basic interest in such measurements clearly ranges from the aspects of pure physics involved in photon-correlation phenomena and the interaction of short pulses with excited atoms to the more-applied considerations involved in understanding the behavior of mode-locked lasers themselves. We have investigated a new technique based on the two-quantum photoeffect which has the promise of great simplicity, quantitative accuracy and general applicability.

The utility of two-quantum processes for picosecond pulse resolution has been demonstrated by a number of investigators with autocorrelation methods involving second harmonic generation¹⁻⁵ and two-photon induced fluorescence.⁶ However, the use of the two-quantum photoelectric effect for this type of measurement has not previously been reported. Further, as reviewed by Weber

and Danielmeyer,⁷ ambiguities arise in the interpretation of the two-quantum correlation experiments without a fairly accurate quantitative knowledge of the intensity autocorrelation function. The width of the central peak of this function is roughly the same for a mode-locked laser pulse as it would be for the normal cw multimode output from the same laser and differences primarily arise in the contrast ratio and finer details of the functional form. We have devised a fairly simple and general technique for the quantitative determination of the second-order intensity correlation function,⁸

$$G^{(2)}(\tau) = \langle I(t)I(t+\tau) \rangle, \quad (1)$$

based on the two-quantum photoeffect. Application of the method to the study of mode-locked argon ion laser pulses has yielded results in reasonable agreement with the theory of Weber and Danielmeyer⁷ and has given experimental agreement on successive time-delay measurements within uncertainties of about ≈ 0.2 picoseconds.

By the two-quantum photoelectric effect we mean the dominant process through which electrons are ejected by a light wave of frequency ν from a photo-surface whose work function W satisfies,

$$h\nu < W < 2h\nu. \quad (2)$$

Here there is no first order photoeffect, and the leading non-zero term in a time-dependent perturbation expansion for the electron emission probability is proportional to the square of the intensity, or in some sense to the fourth power of the total

optical field. Hence the two-quantum photoeffect provides a particularly sensitive method to determine pulse shapes and time delays in the picosecond domain through use of a split-beam apparatus of the type illustrated in Fig. 1.

There are a number of inherent advantages in the use of the two-quantum photoeffect for the present type of experiment: the background noise level from single-photon processes can virtually be eliminated through appropriate choice of the photocathode work function; counting techniques can be incorporated which provide further subtraction of background effects and great linearity; the intrinsic limit on time resolution should be determined by the transit time of the ejected electron over atomic dimensions. Finally, a considerable practical advantage over second harmonic generation arises in the study of low-power mode-locked lasers in the short wavelength part of the visible spectrum where severe phase matching difficulties in the ultraviolet have prevented autocorrelation studies using ^{the} second harmonic generation process.

The present method is illustrated by the schematic diagram in Fig. 1. As with many of the earlier experiments, a chain of mode-locked pulses from a cw laser in the visible spectrum is split between the two arms of a modified Michelson interferometer. In our experiment, one arm of the interferometer is of fixed length and the other is mechanically-varied in length at a continuous rate and a voltage proportional to the variable delay is recorded digitally. After passing through a specially-

designed chopper, the two optical pulses are recombined and focussed on a "uv" photomultiplier tube whose work function W satisfies Eq. (2). The output pulses from the photomultiplier tube are fed through a high-speed integral discriminator to a scaler which is switched between add and subtract modes at the command of a reference voltage provided by the chopper.

Each chopping cycle is divided into four equal time intervals:

- | | |
|---|---------------------|
| Interval (1) -- both beams are combined at the phototube | } counter adds |
| Interval (2) -- neither beam strikes the phototube | |
| Interval (3) -- fixed-delay beam strikes phototube alone | } counter subtracts |
| Interval (4) -- variable-delay beam strikes phototube alone | |

It is readily seen that on the average this procedure subtracts any residual constant background signal, any residual single-quantum counting rate from terms of the type $\langle I(t) \rangle$ or $\langle I(t + \tau) \rangle$; and any double quantum counting rate terms of the type $\langle I^2(t) \rangle$ or $\langle I^2(t + \tau) \rangle$.

If τ were kept constant during the measurement, the double quantum counting rate would include terms analagous to those which produce the fringe pattern in the normal Michelson interferometer. Under our experimental conditions, τ is varied at a small constant rate corresponding to displacements in the variable arm of the interferometer which are large compared to a wavelength over a counting period. Consequently, the "fringe" terms average out to negligibly-small values in the measurement and our net counting rate is closely proportional to the intensity.

correlation function (1). This averaging process presents no significant deleterious effect in the present work since it merely limits the inherent resolution to time intervals $\gg .01$ picoseconds.

In the present experiment, a low power (≈ 100 mW cw) argon ion laser was mode-locked at 5145 \AA using internal acousto-optic loss modulation to provide a pulse repetition frequency of 142 MHz . A phototube was constructed for the experiment consisting of a thin-film molybdenum photocathode attached to a standard Bendix "channeltron" electron multiplier. [The work function corresponds to a cutoff wavelength of about 2900 \AA .] The phototube background counting rate was typically $\approx 5 \text{ Hz}$ and was quite insensitive to the presence of normal room lights in the laboratory. The single-quantum uv counting rate could easily be checked with the 2537 \AA Hg I line and permitted demonstrating that the overall detection system was linear within the statistical fluctuations through counting rates well in excess of 1 MHz . The counting rate obtained when the phototube was exposed directly to the 5145 \AA cw laser beam was typically $\approx 10 \text{ kHz}$ and varied closely as the square of the laser intensity. Very rough measurements indicated that the single-quantum photoefficiency was about 10^{-6} at 2537 \AA . Under the conditions of the experiment, the counting rate at the peak of the autocorrelation function in the mode-locked case out of the difference counter was about 5 kHz . Although the overall efficiency is very low, the absence of any significant background counting rate in the experiment provides

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high signal-to-noise ratios and counting rates which are quite adequate for experimental purposes.

Representative data for the autocorrelation function in the mode-locked case are shown in the upper portion of Fig. 2. In order to deduce the actual width of the mode-locked laser pulse from the autocorrelation data, one must know its functional form. The most commonly-assumed functional forms for the shape of periodic mode-locked lasers are gaussian or lorentzian. These two forms are most convenient for analysis: the folding of a gaussian pulse shape results in a correlation function which is a gaussian with $\sqrt{2}$ greater width; similarly, the folding of a lorentzian is a lorentzian with twice the original pulse width. Beyond mere convenience in the analysis of data, there is some theoretical justification for the assumption of a gaussian pulse shape in the present case: the continuous fourier transform of a gaussian pulse is a gaussian frequency distribution and the laser transition profile is known to be primarily gaussian as a result of Doppler broadening.⁹

Consequently, four-parameter least-squares fits of the data were computed for each run to both the gaussian and lorentzian forms. The four parameters in each case included: a background constant, peak amplitude, location of the peak and full-width at half-maximum response. The gaussian fit (solid line in Fig. 2) was quite good over the entire line in each case examined. The lorentzian fit (dotted line in Fig. 2) was very poor away from the central region of the line in each case examined. In fact,

it was so poor that the lorentzian assumption clearly may be ruled out in the present experiment. Analyses of five separate runs gave an average full width at half-maximum (FWHM) for the original gaussian laser pulse of (78.4 ± 3.8) picoseconds. Although substantially shorter than any previous pulse width reported for mode-locked oscillation on the 5145 \AA Ar II transition, the value of 78 picoseconds is in excellent agreement with direct measurements of the bandwidth of the amplifying transition and with the number of simultaneously oscillating modes observed in the mode-locked condition.

For example, if we assume a gaussian pulse oscillating quasi-sinusoidally at the laser frequency, the power spectrum from a continuous fourier transform would have a FWHM which satisfies

$\Delta \nu_{\text{FWHM}} = 2 \ln_e 2 (\pi \Delta t_{\text{FWHM}})^{-1}$ or a frequency FWHM of 5.6 GHz corresponding to a pulse FWHM of 78 picoseconds. This frequency width is in good agreement with the combined values for the Doppler width and current-broadened lorentz widths for the line obtained under similar discharge conditions,⁹ and would encompass about 40 axial modes in the present laser ($c/2L \approx 142 \text{ MHz}$).

Direct observation of the mode-spectrum with a scanning Fabry-Perot analyzer showed ≈ 43 distinct locked modes oscillating simultaneously under the conditions used to take the correlation data in Fig. 2. As an alternative check, fourier expansion of the quasi-sinusoidal gaussian pulse in the discrete set of uniformly-spaced axial modes gives a power spectrum which is easy to compute numerically. For the pulse width deduced from the data

Greek
"delta"
"nu"
pi

in Fig. 2 (78 picoseconds) a numerical computation of the discrete fourier transform showed that 92.4 percent of the pulse energy would be contained in the first 43 modes centered about the line in the present cavity. A comparison of the mode spectrum seen with the scanning Fabry-Perot and the mode-spectrum computed from the discrete fourier transform of the gaussian pulse is shown in Fig. 3.

The contrast ratios for the autocorrelation data are objectively defined in terms of the fitted parameters for the peak height and the background constant. These ratios were typically in excess of 18:1 for optimum interferometer adjustment, but were also extremely sensitive to the beam alignment.

The data in the lower part of Fig. 2 were taken immediately after the data in the upper portion of the figure, but with the mode-locker turned off. The contrast ratio in that instance was about 1.8:1. The theory of Weber and Danielmeyer⁷ predicts the contrast ratio might approach infinity for a sufficiently large number of coupled oscillating modes and that the ratio should approach 2:1 in the present multimode case without mode-locking.

The least-squares fit parameter determining the peak location also provides a simple method for the measurement of short time delays. Data taken by introducing an optical flat in one arm of the interferometer demonstrated that time intervals ≈ 100 picoseconds could be consistently measured with the present apparatus within discrepancies $\lesssim 0.2$ picoseconds using the least-

squares fitting procedure.

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FIGURE CAPTIONS:

Fig.1. Schematic diagram of the experimental method

used to measure the second-order intensity auto-correlation function, $G^{(2)}(\tau)$.

Greek
'Tau'

Fig.2. Representative intensity auto-correlation data taken

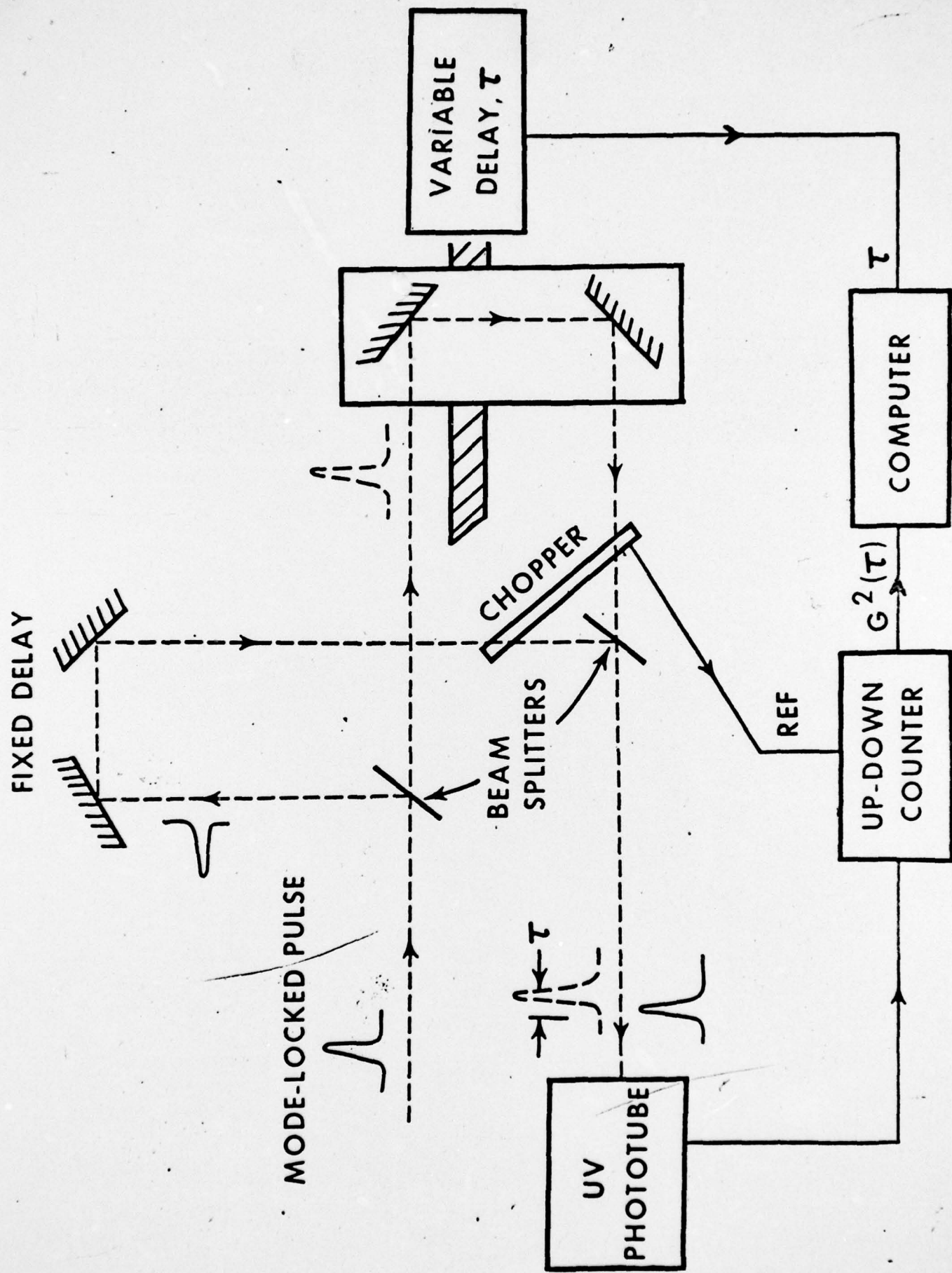
with the apparatus shown in Fig.1. The upper figure shows data taken for a continuous train of mode-locked pulses, together with least-squares fits to gaussian (solid curve) and lorentzian (dotted curve) functional forms.

The full width of the pulse at half-maximum intensity corresponding to the gaussian fit is 78 picoseconds.

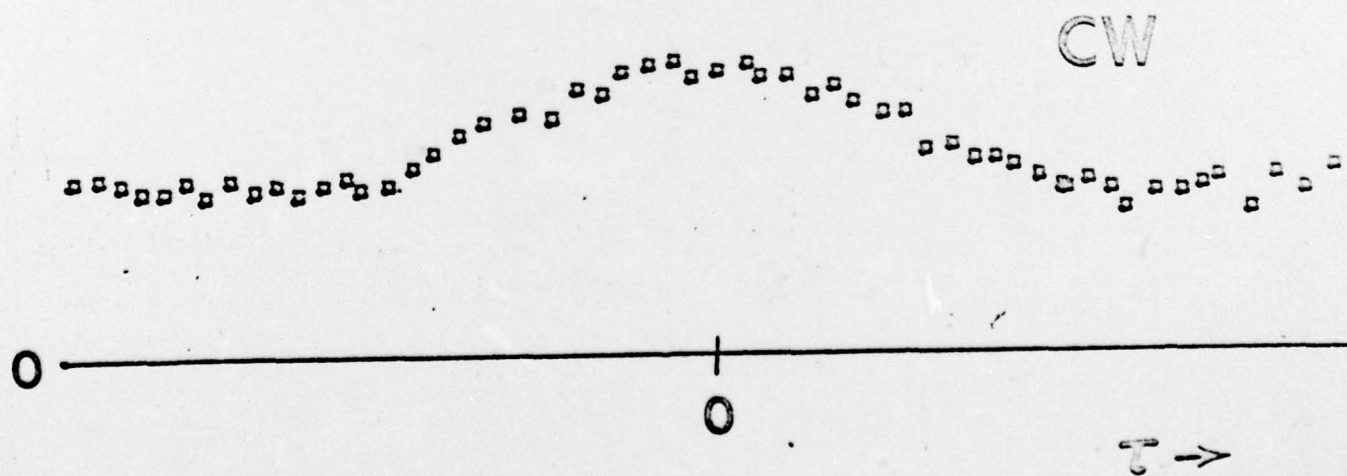
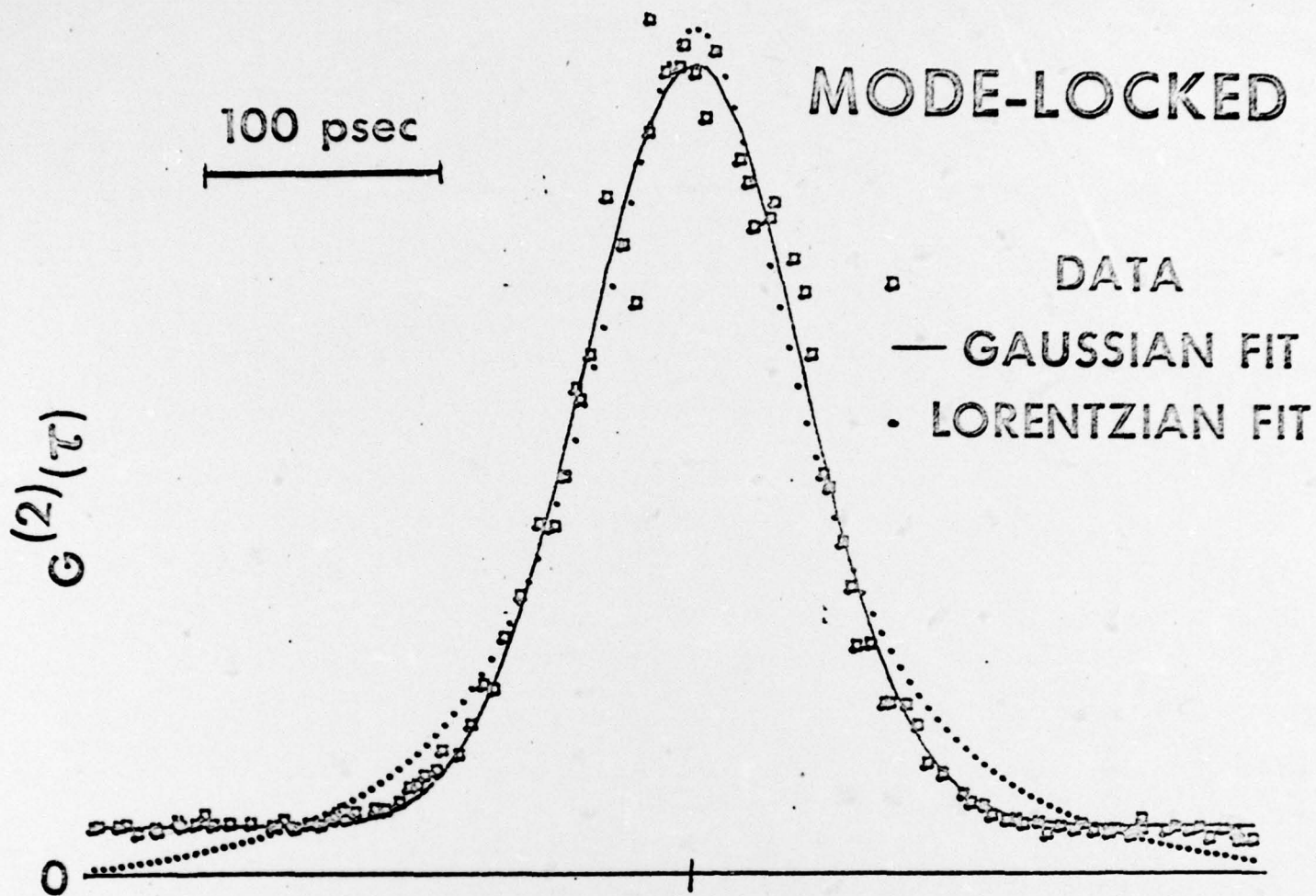
The lower figure shows the auto-correlation data taken with the same apparatus and plotted in the same relative units when the mode-locking element was turned off.

Fig.3. Power spectrum of locked modes characteristic of the

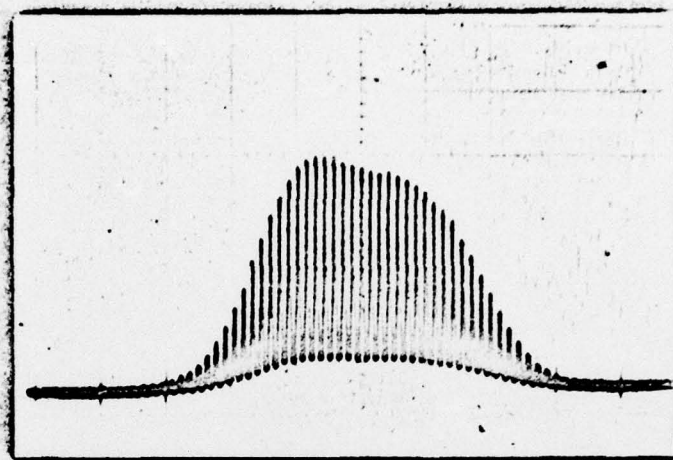
laser used ($c/2L = 142$ MHz): (a) observed experimentally with a scanning Fabry-Perot interferometer; (b) computed from the correlation data in Fig.2 by expanding the corresponding quasi-sinusoidal gaussian envelope pulse in the set of discrete cavity modes uniformly-spaced at $c/2L$.



55%



(b)



(c)

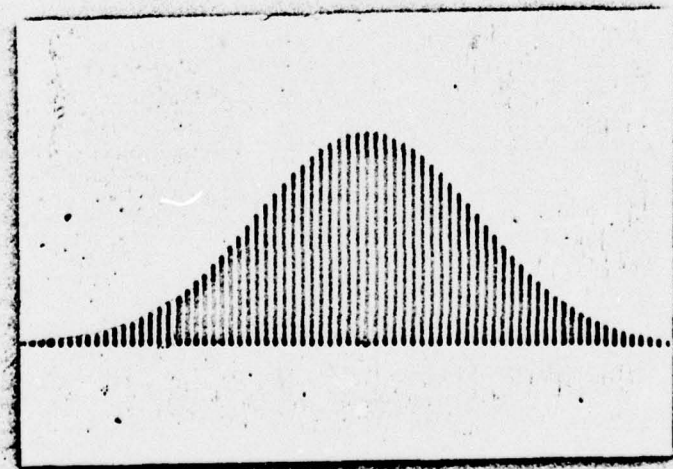


Fig. 3 Bennett, Carlin, Gills